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[Title of the Invention] APPARATUS AND PROCESS FOR SYNTHESIS OF CARBON NANOTUBES OR CARBON NANOFIBERS USING FLAMES

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[Purpose]	The above application is filed in accordance with Articles 42 and 60 of Korean Patent Law.

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[Abstract of the Disclosure]

[Abstract]

Disclosed herein is an apparatus for synthesizing a carbon nano-material comprising: a source supplier for supplying a reaction gas and a metallic catalyst in isolation from atmospheric condition; a reactor communicating with the source supplier, wherein synthesis of the carbon nano-material occurs by the reaction gas and the metallic catalyst; a flame providing means for providing flames outside the reactor for maintaining the reactor at a temperature proper for the synthesis of the carbon nano-material; and a collecting means for collecting the carbon nano-material from products generated in the reactor.

The apparatus for synthesizing carbon nano-materials of the present invention is advantageous in that the apparatus is simply configured and can be operated at an inexpensive cost by providing an environment for synthesis of the carbon nano-material by means of indirect heating using flames.

[Representative drawing]

Fig. 2

[Keyword]

carbon nanotube, carbon nanofiber, carbon nano-material, flame, burner, electrostatic precipitation

[Specification]

[Title]

APPARATUS AND PROCESS FOR SYNTHESIS OF CARBON NANOTUBES OR CARBON NANOFIBERS USING FLAMES

[Brief Description of the Drawings]

Fig. 1a shows a schematic of a prior art arc discharging device.

Fig. 1b shows a schematic of a prior art arc generating device.

Fig. 1c is a plan view of the arc generating device shown in Fig. 1b.

Fig. 2 shows a schematic of an apparatus for synthesizing carbon nano-materials according to a first embodiment of the present invention.

Fig. 2a shows a schematic of a metallic catalyst supplier of Fig. 2.

Fig. 3 shows a schematic of an apparatus for synthesizing carbon nano-materials according to a second embodiment of the present invention.

Fig. 3a illustrates a schematic of a reaction gas supplier and a metallic catalyst supplier of Fig. 3.

Fig. 4 shows a schematic of an apparatus for synthesizing carbon nano-materials according to a third embodiment of the present invention.

Fig. 5 shows a schematic of an example of a surface flame burner used in the present invention.

Fig. 6 is a schematic of a collector of the present invention.

<Description of Legend in the Drawings>

60: reaction gas supplier

62: metallic catalyst supplier

64: Fuel and oxidizer supplier

66: burner

68: reflector

70: reactor

72: heat exchanger

74: collector

[Detailed Description of the Invention]

[Object of the Invention]

[Field of Invention and Prior art of the Field]

The present invention relates to apparatus and process for synthesizing carbon nano-materials and, more particularly, to an apparatus for synthesizing carbon nano-materials, such as carbon nanotubes or carbon nanofibers, wherein the carbon nano-material is synthesized by means of indirect heating using flames, and a synthesizing process using such an apparatus.

A carbon nanotube or a carbon nanofiber is composed of a plurality of cylindrically rolled graphite sheets, wherein a plurality of hexagonal ring shapes having one carbon atom and three carbon atoms joined thereto with sp2 bonding are repeated in a honeycomb form. The diameter of such a cylindrical shape ranges from several nanometers to a hundred nanometers and the length thereof is a dozen times through a thousand times as long as the diameter.

Carbon nanotubes may be classified into a single-wall nanotube, a multi-wall nanotube, and a rope nanotube, in terms of the form of the rolled graphite sheets. Further, they have various electrical characteristics that are determined according to the roll angle and shape of the graphite sheet. For example, it has been known that carbon nanotubes or carbon nanofibers have an electrical conductivity such as that of metal when they are in an armchair configuration. Further, it has been known that carbon nanotubes or carbon nanofibers have the characteristic of a semiconductor when being in a zig-zag configuration.

Such carbon nanotubes or carbon nanofibers are chemically stable with excellent electrical characteristics and high mechanical strength. Therefore, they are expected to be widely applied in the information and technology industry in a variety of manners.

Prior art apparatus for synthesizing such carbon nanotubes are shown in Figs. 1a to 1c. Fig. 1a shows a rotary electrode arc-discharging device. Fig. 1b shows an arc generating device. Fig. 1c is a plan view of the arc generating device.

As shown in Fig. 1a, the rotary electrode arc-discharging device comprises the following: a vacuum vessel 13 in which high vacuum is possible through a vacuum exhaust port 11; an insulation chamber 17 positioned inside the vacuum vessel 13 and being insulated by an insulator 15 from the outside; a low temperature liquid injection port 19, through which low-temperature liquid such as liquid nitrogen, liquid helium or ultra low-temperature water can be injected, the liquid injection port being positioned at a right top side of the chamber 17; and an opening and closing aperture 21, through which the arc-generating device (*see* Fig. 1b) is inserted into the chamber 17.

Further, the prior art rotary electrode arc-discharging device is provided with the following: an outlet 23 positioned at a bottom of the chamber 17 for discharging the low temperature liquid or the synthesized carbon nanotubes therethrough; a discharging valve 25 for controlling the low temperature liquid or the synthesized carbon nanotubes discharged through the outlet 23; and view ports 27a, 27b positioned at a right side of the vacuum vessel 13 and at a right side of the chamber 17, respectively, for allowing observing the inside of the chamber 17.

Figs. 1b and 1c show the arc generating device. The arc generating device includes: a cover portion 31 adapted to be fitted to the above-described opening and closing aperture 21; a cathode holder 33 extending into the chamber 17 as passing through the cover portion 31 at a side thereof; a cathode holder support 35a, 35b coupled to the cathode holder 33 and

being movable up and down; and a cathode 37 made from graphite, the cathode being connected to the cathode holder support 35a, 35b and applying negative voltage. Further, the arc generating device includes: a position adjusting portion 41 for adjusting a vertical position of the cathode, the position adjusting portion being apart from the cathode holder 33 and being connected to the cathode holder support 35a, 35b; an anode support 43 being apart from the position adjusting portion 41 and extending into the chamber 17 as passing through one side of the cover portion 31; a chuck 45 connected to the high-speed rotatable anode support 43; and an anode 47 connected to the chuck 45 opposite the cathode 37 for applying positive voltage, the anode being rotatable at high speed by means of the anode support 43 and the chuck 45.

However, the prior art carbon nanotube synthesizing apparatus is disadvantageous in that it has high manufacturing costs since it uses electrical energy as a heat source supplying heat necessary for synthesis reaction. Further, it is also disadvantageous in that the constitution of the arc generating device is complex.

[Technical Problem Solved by the Invention]

The present invention has been made to solve the above problems. It is an object of the present invention to provide an apparatus for synthesizing carbon nano-materials, which can be configured and operated at an inexpensive cost by supplying heat necessary for synthesis of carbon nano-materials in a heating manner using flames.

The above and other objects are accomplished with a an apparatus for synthesizing carbon nano-material, comprising: a source supplier for supplying a reaction gas and metallic catalyst in isolation from atmospheric condition; a reactor communicating with the source supplier, wherein synthesis of the carbon nano-material occurs by the reaction gas and the metallic catalyst; a flame providing means for providing flames outside the reactor for maintaining the reactor at a temperature proper for the synthesis of the carbon

nano-material; and a collecting means for collecting the carbon nano-material from products generated in the reactor.

Further, the present invention provides a process for synthesizing carbon nano-material using the above-described apparatus.

[Constitution of the Invention]

Preferred embodiments of the present invention will now be described with reference to the accompanying drawings.

The term "carbon nano-material," used throughout the description, represents materials containing carbon, with a diameter of several nanometers through a hundred nanometers, such as carbon nanotubes and carbon nanofibers.

Fig. 2 is a schematic of an apparatus for synthesizing carbon nano-materials in accordance with a first embodiment of the present invention. The apparatus for synthesizing carbon nano-materials comprises: reaction gas supplier 60, metallic catalyst supplier 62, reflector 68, reactor 70, burner 66, heat exchanger 72, and collector 74.

Reaction gas supplier 60 serves to supply to reactor 70 the carbon source gas necessary for synthesis of the carbon nano-material. Reaction gas supplier 60 is connected to main supply tube 78, which communicates with the reactor 70, via supply tube 78a. Gaseous hydrocarbons, such as methane, ethylene, acetylene, cyclohexane, benzene, xylene, etc. are used as carbon source gases. Gas cylinder for storing hydrocarbon under pressure may be used as reaction gas supplier 60.

Metallic catalyst supplier 62 serves to supply gaseous metallic catalyst necessary for synthesis of the carbon nano-material to reactor 70. Metallic catalyst supplier 62 is connected to main supply tube 78 via a supply tube 78b. Metal nitrate such as $Fe(No_3)_3$, $Ni(NO_3)_2$ is used as the metallic catalyst. Organic metallic compound such as $Fe(CO)_5$, $Mo(CO)_6$, $CO_2(CO)_8$, $(C_5H_5)_2Fe$ and $Ni(CO)_5$ is available as the metallic catalyst source. In

case of source material that is hardly evaporated by only an evaporator, e.g., $(C_5H_5)_2Fe$, a separate sublimer (not shown) may be used to help its evaporation for the metallic catalyst in gaseous state.

As shown in Fig. 2a, metallic catalyst supplier 62 may be embodied with a carrier gas supplier 62b and evaporator 62a containing the metallic catalyst source material. Evaporator 62a is connected to carrier gas supplier 62b through flow rate control valve 62c. Metallic catalysts in a solid or liquid state are accommodated in evaporator 62a as the metallic catalyst source material and are heated for evaporation by heater 96 positioned in a lower portion of evaporator 62a. Heater 96 may be embodied with "a hot plate," for example. Flow rate control valve 62a functions to adjust the flow rate of the carrier gas being supplied to evaporator 62a. Inert gas such as argon (Ar) may be employed as the carrier gas. In case argon gas is employed as the carrier gas, carrier gas supplier 62b may be embodied with a general gas cylinder that contains argon under pressure. The metallic catalyst evaporated in evaporator 62a is carried toward main supply tube 78 by the carrier gas.

Gases supplied from reaction gas supplier 60 and metallic catalyst supplier 62 are mixed in main supply tube 78 to be fed to reactor 70. Main supply tube 78 and reactor 70 are preferably made of quartz.

In a first embodiment of the present invention, reactor 70 extends in a helical shape. The helical shape of reactor 70 enables more portions of reactor 70 to be exposed to a flame provided by burner 66. As a result, the reaction gas and the metallic catalyst are put under an environment for synthesis of carbon nano-materials, for a longer period of time, by traversing helical shaped reactor 70.

In the first embodiment, burner 66 is mounted under reactor 70, while reflector 68 is mounted above reactor 70.

Burner 66 serves to heat reactor 70 to maintain an optimal temperature in reactor 70, at which much carbon nano-material is synthesized from the carbon source gas and the metallic catalyst. Fuel and oxidizer supplier 64 supplies fuel and oxidizer, which are needed for combustion, to burner 66 through supply tube 64a. Preferably, fuel whose quantity of heat can be easily controlled is used. In particular, LNG or LPG is preferable. Oxygen is the preferable oxidizer.

The quantity of heat provided by burner 66 has to be finely adjusted in order to form the optimal temperature in reactor 70. The quantity of heat may be adjusted by adjusting the amount of the fuel and the oxidizer being supplied from fuel and oxidizer supplier 64 or by changing the distance between burner 66 and reactor 70. In order to change the distance between burner 66 and reactor 70, it is preferable that burner 66 is movable in a direction indicated by the arrow, so that burner 66 and reactor 70 get closer to or get more distant from each other. In the first embodiment, since reactor 70 has a helical shape close to a circular shape, burner 66 should preferably have a circular cross-section to result in a circular shaped flame. Examples of commercial burners appropriate to the present invention will be discussed later in detail with reference to Fig. 5.

Reflector 68 is positioned opposite to burner 66 about reactor 70 to reflect heat provided by burner 66 toward reactor 70. Reflector 68 is preferably movable in a direction indicated by the arrow like burner 66, so that the distance between reflector 68 and reactor 70 can be changed.

Heat exchanger 72 cools the synthesized carbon nano-material escaping from reactor 70. A water-cooling heat exchanger using water as cooling media is preferred. The use of heat exchanger 72 may be optional. In case the produced carbon nano-material has a temperature appropriate to the processes in collector 74 at the time of its arriving at collector 74, heat exchanger 72 may be unnecessary. In particular, when supply tube 78c

communicating with reactor 70 has sufficient length, a separate heat exchanger 72 is not necessary since the products of the carbon nano-material are cooled in the course of traveling.

Collector 74 collects the products of the carbon nano-material. Collector 74 of the present invention collects the carbon nano-materials using electrostatic precipitation. Detailed description about collector 74 will be given later with reference to Fig. 6. However, according to the present invention, a paraffin bubbling device may be used as a collector for collecting the carbon nano-materials.

Fig. 3 shows a schematic of an apparatus for synthesizing carbon nano-materials in accordance with a second embodiment of the present invention.

In description on the second embodiment, like parts or components with those shown in the first embodiment are designated with same reference numerals and description for those will be omitted.

Unlike the first embodiment where the reaction gas and the metallic catalyst are separately supplied to and mixed in main supply tube 78, in the second embodiment, the reaction gas is directed to metallic catalyst supplier 63 via supply tube 78a. In such a case, as shown in Fig. 3a, metallic catalyst supplier 63 may be embodied with only an evaporator and reaction gas supplier 60 may be embodied with a gas cylinder for storing the reaction gas under pressure. In metallic catalyst supplier 63, the metallic catalyst in a gaseous state is generated from the metallic catalyst source in a liquid state through evaporation and mixed with the reaction gas supplied from reaction gas supplier 60. The reaction gas functions as the carrier gas that carries the mixed gases to main supply tube 78.

Fig. 4 shows an apparatus for synthesizing carbon nano-materials in accordance with a third embodiment.

In description on the apparatus of the third embodiment, like parts or components with those shown in the first embodiment are designated with like reference numerals and

description for those will be omitted.

In the third embodiment, reactor 70' extends in a zigzag form and is made of quartz. Pair of burners 66' are provided above and under reactor 70'. It is preferable that burners 66' are identical in shape and have a rectangular shape capable of covering the whole area of zigzag reactor 70'.

Further, it is preferable that burners 66' are movable, so that burners 66' get closer toward or more distant from reactor 70'. With this configuration, the quantity of heat to be provided to reactor 70' may be easily adjusted.

Fig. 5 illustrates one example of a burner 100 to be used as burner 66, 66' of the present invention. Preferably, flames, which are used to heat reactor 70, 70' at a required temperature, are pre-mixed flat flames or partially pre-mixed flat flames that ensure good radiant heat transfer and generate less impurities. As shown in Fig. 5, surface flame burner 100 is appropriated as a burner for providing such flames.

Surface flame burner 100 is provided with main body 108 and mat 104. As shown by the arrow, a mixture of fuel gas and oxidizer (or, oxygen) is introduced from a central lower portion of main body 108, at a constant flow speed. The mixture is burnt in the course of passing through gas permeable mat 104. In combustion, length h of the flame, which can depend on the flow speed of the mixture, is approximately 1cm. Mat 104 is made of a metal fiber with porosity. Various commercial mats can be applied to the present invention.

Further, as various commercial burners are known, those skilled in the art will recognize that any type of burner capable of providing the flat flames is applicable to the apparatus of the present invention.

Fig. 6 depicts one example 80 of a collector using an electrostatic precipitating method according to the present invention. Collector 80 is provided with charging unit 82

and separation unit 84. In charging unit 82, a streamer of plasma having low temperature is established. Large amounts of ions are generated in the streamer by applying an alternating current provided by AC power source 82a. When carbon nano-material 92, synthesized in reactors 70, 70', arrives at charging unit 82, it is positively or negatively charged by distributed ions 90.

In separation unit 84, a direct-current electric field is established between a pair of collecting plates 86. Collecting plates 86 are connected to DC power source 84a, and, therefore, have different electric polarities from each other. When charged carbon nano-material 92 arrives at separation unit 84 after leaving charging unit 82, it is attracted to collecting plate 86 that has a polarity opposite to its own polarity and adheres thereto. Next, carbon nano-material 92, adhered to collecting plate 86, is scratched from collecting plate 86 and then purified through a filter.

[Effect of the Invention]

According to the present invention, the apparatus for synthesizing carbon nano-materials may be configured at a more reasonable price, when compared to the prior art carbon nano-material synthesizing apparatus using electric energy, by providing an environment for carbon nano-material synthesis, which is performed under indirect heating using flames.

Further, since the space in which the synthesis of the carbon nano-material occurs and the space in which the combustion by the burner occurs are closed off from each other, impurities generated by the flames will not contaminate the products.

Further, the apparatus for synthesizing carbon nano-materials of the present invention can be operated continuously without work such as exchanging carbon electrodes, exchanging a substrate, etc. Therefore, the apparatus for synthesizing carbon nano-materials of the present invention is suitable for mass production of carbon nano-materials.

What is claimed is:

1. An apparatus for synthesizing a carbon nano-material, comprising:

a source supplier for supplying a reaction gas and a metallic catalyst in isolation from atmospheric condition;

a reactor communicating with the source supplier, wherein synthesis of the carbon nano-material occurs by the reaction gas and the metallic catalyst;

a flame providing means for providing flames outside the reactor for maintaining the reactor at a temperature proper for the synthesis of the carbon nano-material; and

a collecting means for collecting the carbon nano-material from products generated in the reactor.

2. The apparatus of Claim 1, wherein the reaction gas is selected from a group including methane, ethylene, acetylene, carbon monoxide, cyclohexane, benzene, and xylene.

3. The apparatus of Claim 1, wherein the metallic catalyst is metal nitrate.

4. The apparatus of Claim 1, wherein the reactor is a tube made of quartz.

5. The apparatus of Claim 1, wherein the flame providing means is a surface flame burner.

6. The apparatus of Claim 5, wherein the surface flame burner is disposed in a vertically symmetrical relation about the reactor.

7. The apparatus of Claim 1 or 4, wherein the reactor extends in a helical form.

8. The apparatus of Claim 1 or 4, wherein the reactor extends in a zigzag form.

9. The apparatus of Claim 1, wherein the collecting means comprises:

a charging unit communicating with the reactor, wherein produced carbon nano-material is electrically charged by an applied alternative current electrical field; and

a separation unit communicating with the charging unit and including a pair of plates generating a direct-current electric field, wherein the produced carbon nano-material adheres to each one of the pair of plates.

10. A process for synthesizing a carbon nano-material using an apparatus for synthesizing a carbon nano-material according to any one of Claims 1 to 6 and 9.

11. A process for synthesizing a carbon nano-material using an apparatus for synthesizing a carbon nano-material according to Claim 7.

12. A process for synthesizing a carbon nano-material using an apparatus for synthesizing a carbon nano-material according to Claim 8.

Fig. 1a

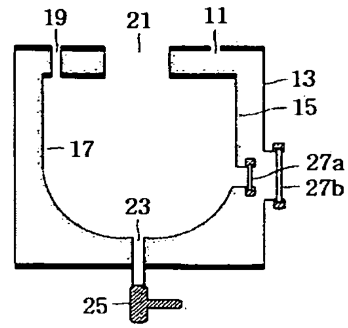


Fig. 1b

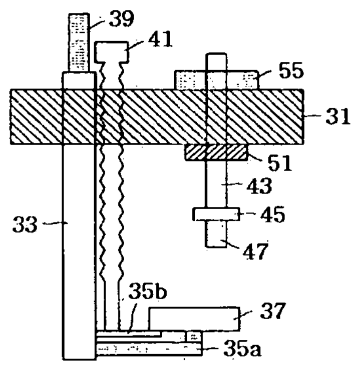


Fig. 1c

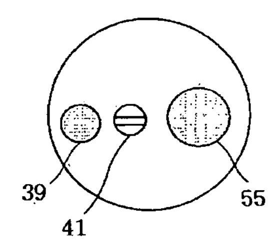
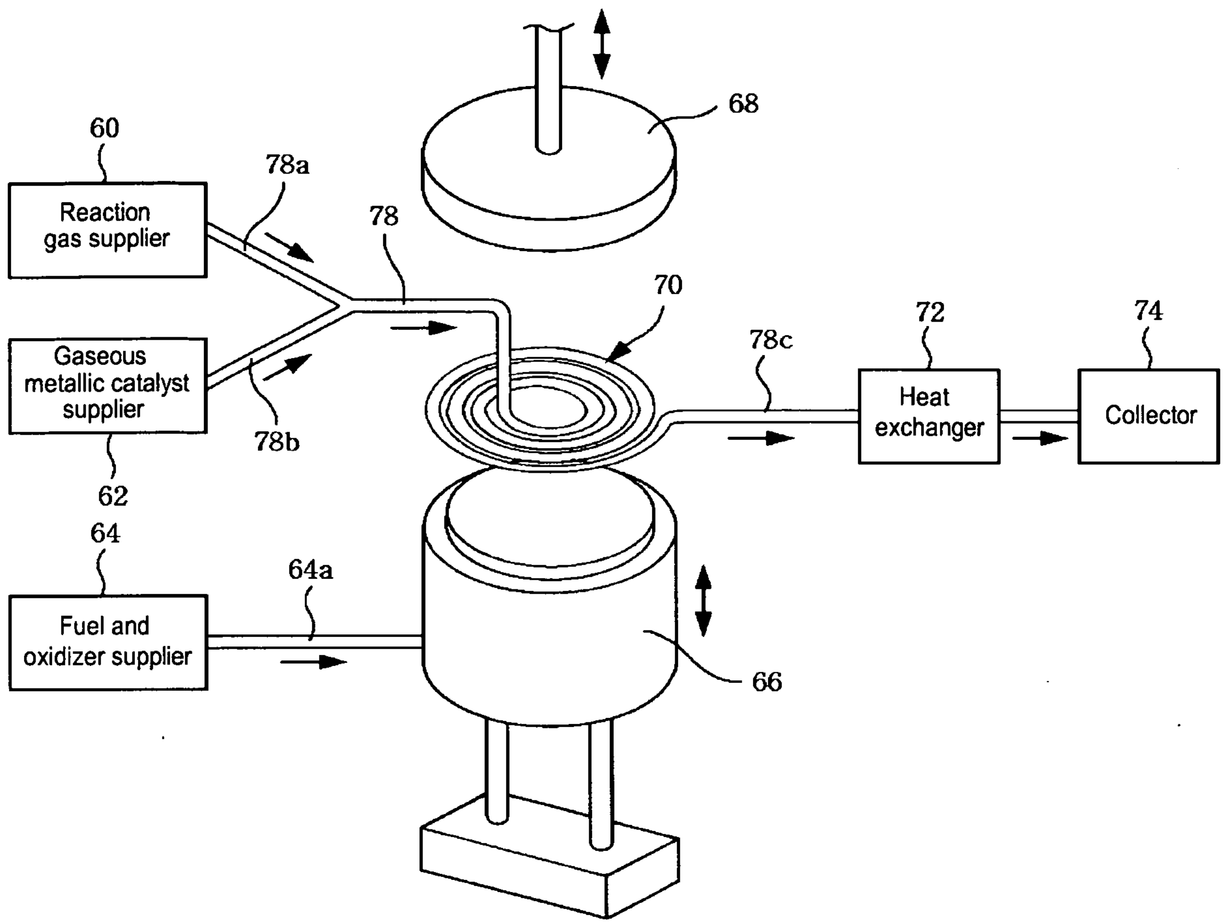


Fig. 2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60
78a
Reaction gas supplier
78
70
72
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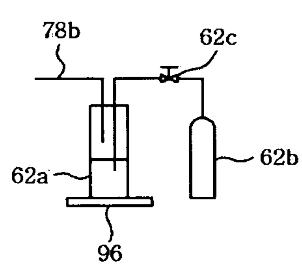
Gaseous metallic catalyst supplier
78b
Heat exchanger
Collector
62
64
64a
Fuel and oxidizer supplier
66

Fig. 2a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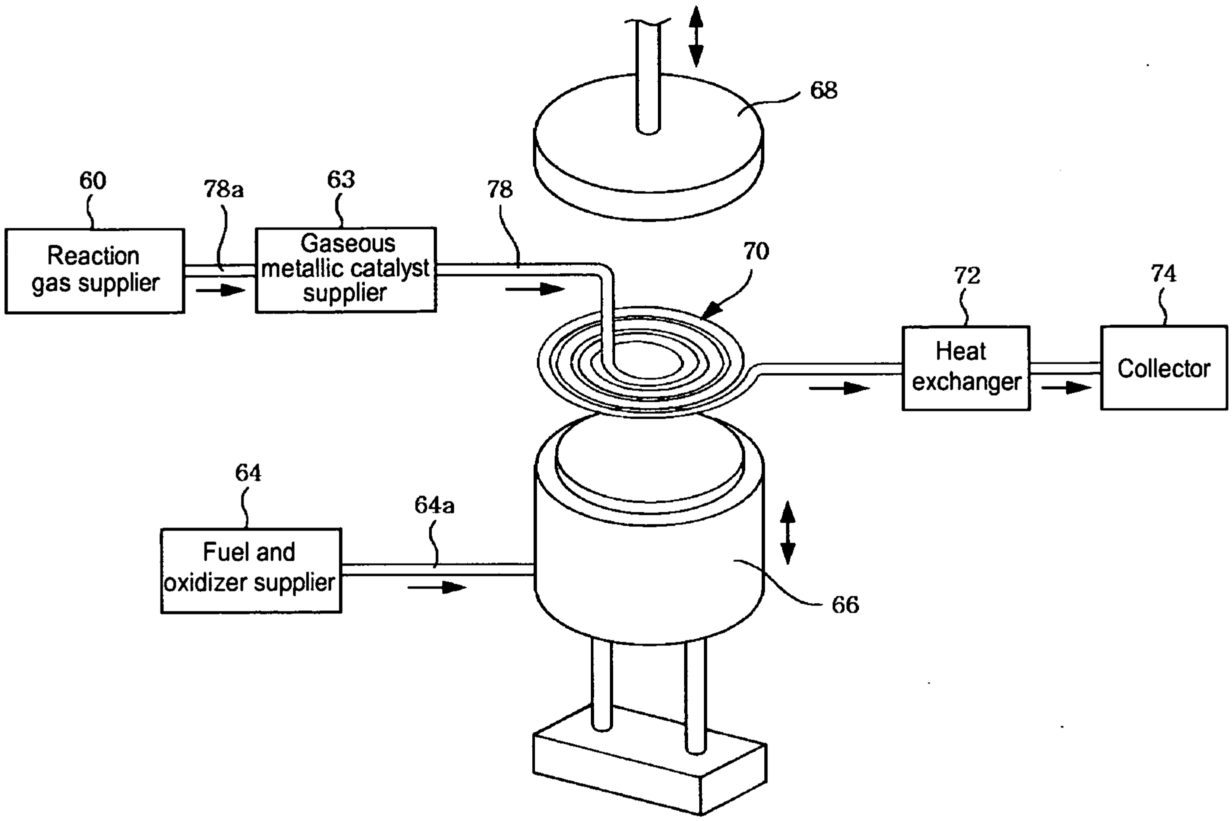
62c
62a
62b
96

Fig. 3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60
78a
63
78
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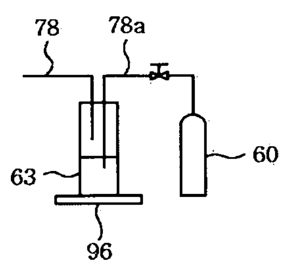
Gaseous metallic catalyst supplier
70
72
74
Heat exchanger
Collector
64
64a
Fuel and oxidizer supplier
66

Fig. 3a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60
63
96

Fig. 4

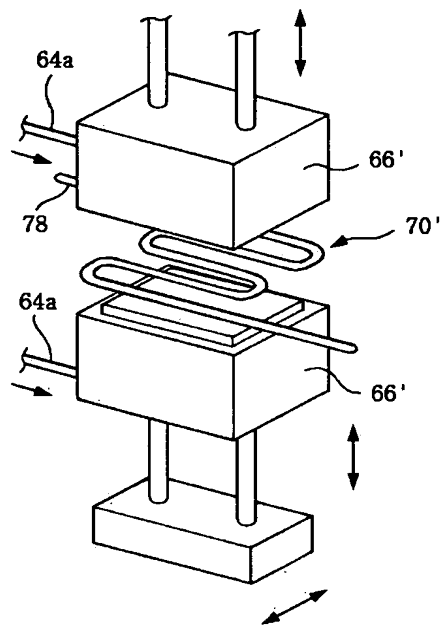
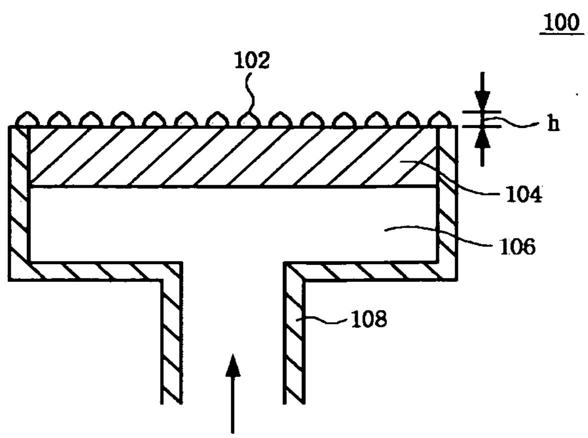


Fig. 5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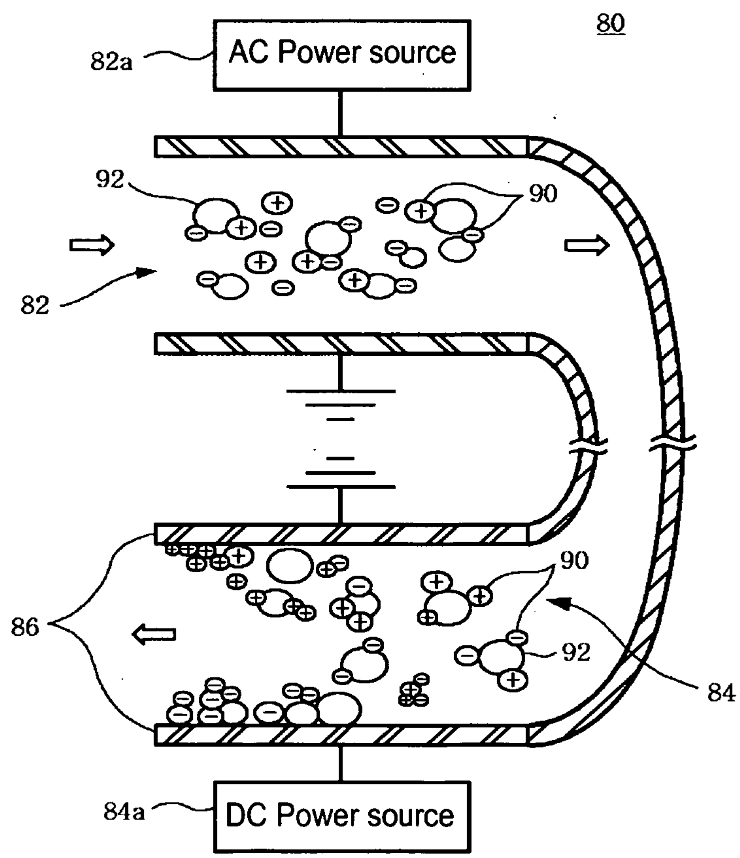
102
h
104
106
108

Fig. 6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92
90
82
86
90
92
84
84a
DC Power source